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to Measure Plutonium Mass and Multiplication**

J. K. Mattingly

J. S. Neal

J. T. Mihalcz

**Y-12
National
Security
Complex**

**Nuclear Materials Management and
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TIME-DEPENDENT COINCIDENCE METHOD TO MEASURE PLUTONIUM MASS AND MULTIPLICATION

John Mattingly, John Neal, and John Mihalcz
Oak Ridge National Laboratory
P.O. Box 2008 MS6004, Oak Ridge, Tennessee

INTRODUCTION

Future nuclear disarmament agreements between nations may require technical measures to ascertain each participating nation's adherence to the agreement. Almost certainly, measurement technologies and analytical methods will have to be developed by the participating nations *jointly*. In this way each participant has both *confidence* in the technology's efficacy and *trust* in its implementation.

With the support of the National Nuclear Security Administration's Office of Nonproliferation Policy (NNSA NA-241), the Oak Ridge National Laboratory (ORNL) and the All-Russian Scientific Research Institute of Experimental Physics (VNIIEF) have taken first steps to jointly develop and implement a radiation measurement technique to inspect plutonium.

In June and July 2000, personnel from ORNL and VNIIEF performed joint experiments on unclassified plutonium metal (δ -phase, 1.77%- ^{240}Pu) spherical shells at VNIIEF facilities in Sarov, Russia [1,2]. The measurements were performed using the Nuclear Materials Identification System (NMIS). The subsequent analysis demonstrates how NMIS can be applied to passively measure the mass and multiplication of plutonium spherical shells.

DESCRIPTION OF WORK

In passive mode¹, an NMIS measurement is similar to a Rossi- α measurement. An array of fast plastic scintillators is used to accumulate the distribution of coincident detector counts. This coincidence-distribution is the rate of real coincidence (i.e., total coincidence less accidental coincidence) as a function of the time-delay between detectors (at zero time-delay, the detectors' signals are synchronized).

The NMIS was applied in its passive mode to eight plutonium spherical shells. The shells' properties spanned the following ranges:

- Composition: δ -phase plutonium-metal, constant
- Relative ^{240}Pu -content ($f_{240\text{Pu}}$): $f_{240\text{Pu}} = 1.77\%$ {g ^{240}Pu / g Pu}, constant
- Inner radius (r_1): $10.0 \text{ mm} \leq r_1 \leq 53.5 \text{ mm}$, mean $r_1 = 33.5 \text{ mm}$
- Outer radius (r_2): $31.5 \text{ mm} \leq r_2 \leq 60.0 \text{ mm}$, mean $r_2 = 46.6 \text{ mm}$
- Radial thickness (Δr): $6.4 \text{ mm} \leq \Delta r \leq 30.2 \text{ mm}$, mean $\Delta r = 13.1 \text{ mm}$
- Plutonium mass (m_{Pu}): $1829 \text{ g} \leq m_{\text{Pu}} \leq 4468 \text{ g}$, mean $m_{\text{Pu}} = 3265 \text{ g}$

¹ The NMIS also operates in an active mode where it uses a neutron source to induce fission in the target. In active mode, an NMIS measurement is analogous to simultaneous pulsed-neutron and Rossi- α measurements.

The measurements were analyzed to extract the attributes of each plutonium shell. Given that the samples measured were of constant composition, geometry, and relative ^{240}Pu -content², each shell is completely described by any two of the following four properties:

- Inner radius r_1
- Outer radius r_2
- Mass m , either ^{239}Pu mass $m_{^{239}\text{Pu}}$, ^{240}Pu mass $m_{^{240}\text{Pu}}$, or total Pu mass m_{Pu} ³
- Radial thickness Δr

Of these, generally only *mass* is considered an attribute of interest; the second property (whichever is chosen) can be considered to be a parameter of the attribute-estimation procedure, much as multiplication

$$M = \frac{1}{1 - k_{\text{eff}}}$$

is a parameter necessary to accurately estimate mass via most neutron measurements.

In order to accurately account for multiplication effects in the interpretation of the measurements, Monte Carlo calculations were performed to estimate the unreflected k_{eff} of hypothetical spherical shells whose dimensions span those of the VNIIEF shells. Furthermore, the span of relative ^{240}Pu -content was expanded to $1\% \leq f_{^{240}\text{Pu}} \leq 20\%$. The approximate relationship between total plutonium mass m_{Pu} , radial thickness Δr , relative ^{240}Pu -content $f_{^{240}\text{Pu}}$, and multiplication factor k_{eff} was estimated via least-squares regression as:

$$k_{\text{eff}} \approx 0.034 \frac{m_{\text{Pu}}^{0.156} \Delta r^{0.465}}{f_{^{240}\text{Pu}}^{0.015}}$$

Observe that over this span ($0.14 \leq k_{\text{eff}} \leq 0.89$), the effect of relative ^{240}Pu -content is almost negligible.

The features of the coincidence-distribution used to estimate the multiplication parameter and mass attribute are derived from its width and integral. For this study, the distribution width was taken as its full-width at tenth-maximum, or FWTM. The distribution's integral was taken as the area beneath the FWTM, a.k.a., its FWTM area. Subsequently, the width and integral will respectively be denoted by W_{FWTM} and A_{FWTM} .

RESULTS

The width and integral features were analyzed to model their dependence upon multiplication and mass. The width tends to grow with increasing multiplication following a logarithmic trend, and the integral tends to grow with the product of multiplication and mass following a power

² Only 1.77%- ^{240}Pu shells were available at VNIIEF. This is unfortunate because without variation in the relative ^{240}Pu -content, it cannot be demonstrated from these measurements alone that NMIS is capable of extracting this attribute. Future joint measurements planned between the All-Russian Scientific Research Institute of Theoretical Physics (VNIITF), VNIIEF, and ORNL will demonstrate this capability through additional measurements of 10%- ^{240}Pu shells at VNIITF in Snezhinsk, Russia.

³ Due to the constant relative ^{240}Pu -content, ^{239}Pu mass, ^{240}Pu mass, and total plutonium mass are directly proportional and are therefore equivalent measures of mass for these particular spherical shells.

rule. Least-squares regression quantifies these observations. The width feature is approximately proportional to multiplication according to

$$W_{FWTM} \approx 12.53 \ln M + 22.13 \quad (1)$$

and the integral feature is approximately proportional to the product of multiplication and mass according to

$$A_{FWTM} \approx 2.37 (M \cdot m_{240Pu})^{1.14} \quad (2)$$

This *forward* model ((1) and (2)) is readily solved to yield the *inverse* model that estimates multiplication and mass given the width and integral features:

$$M \approx \exp\left(\frac{W_{FWTM} - 22.13}{12.53}\right) \quad (3a)$$

$$m_{240Pu} \approx \left(\frac{A_{FWTM}}{2.37}\right)^{1/1.14} \exp\left(-\frac{W_{FWTM} - 22.13}{12.53}\right) \quad (3b)$$

Substituting a measured width and integral into the preceding inverse model yields an estimate of multiplication and mass. The inverse model applied to the width and integral features measured for all the VNIIEF shells yields fairly accurate estimates of mass and multiplication for each shell. In a root-mean-square (RMS) sense, the inverse model estimates multiplication to within 5% of its actual value and mass to within 7% of its actual value.

In its application to an unknown plutonium-metal spherical shell, the FWTM and FWTM area of the coincidence-distribution acquired from a passive NMIS measurement would be substituted into the inverse model to obtain an estimate of the ^{240}Pu mass according to (3b). If the unknown shell falls within the span $(1.2 \leq M \leq 9.1) \times (18 \text{ g} \leq m_{240Pu} \leq 894 \text{ g})$ then the relative error of the estimated ^{240}Pu mass can be expected to be less than 10%. Consequently, this jointly developed method shows promise as a means to inspect plutonium for domestic and international safeguards and disarmament agreement verification. Further joint experiments will expand its applicability.

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